Role of surface anisotropy for magnetic impurities in electron dephasing and energy relaxation and their size effect

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Recently the electron dephasing and energy relaxation due to different magnetic impurities have been extensively investigated experimentally in thin wires and in this Letter these quantities are theoretically studied. It was shown earlier that a magnetic impurity in a metallic host with strong spin-orbit interaction experiences a surface anisotropy of the form $H = K_d(\mathbf{nS})^2$ which causes size effects for impurities with integer spin. Here we show that the dephasing and the energy relaxation are influenced by the surface anisotropy in very different ways for integer spin having a singlet ground state. That must result also in strong size effects and may resolve the puzzle between the concentrations estimated from the two kind of experiments.

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In the present Letter, to our knowledge, is the first time a mechanism is presented which can resolve the seriously puzzling observation that in some cases the influence of magnetic impurities on electron dephasing and energy relaxation have drastically different strengths. The surprising difference in the estimated impurity concentration raised the doubt about the role and even the presence of magnetic impurities [1]. That mechanism is based on surface magnetic anisotropy [2] which is resulting in a strong size dependence.

The size dependence of the Kondo effect [3, 4] was discovered more than ten years ago and since than it has been carefully studied experimentally [5, 6, 7, 8, 9, 10]. That cannot be attributed to the size of the Kondo screening cloud reduced by the size of the sample as only the energy separation of the metallic electron levels are relevant. That problem was resolved by the suggestion that the magnetic impurities in the metallic host with strong spin-orbit interaction experience a surface anisotropy [2].

In mesoscopic metallic systems the electron dephasing and energy relaxation are the central issues in understanding their transport properties [11]. The interest has been intensified by the debate over the saturation of dephasing at low temperature [12]. The dephasing is determined e.g. from measurements of magnetoresistance and Aharonov-Bohm rings in magnetic field [12, 13, 14, 15] while the energy relaxation from transport in short wires is found by determining the nonequilibrium electron energy distribution as shown by the Saclay group [16]. In many cases the deviations from the expectations of the theory of Altshuler, Aronov and coworkers [17] were attributed to the presence of magnetic impurities either implanted or contained by the starting material as contamination. In addition to the energy relaxation due to electron-electron, electron-phonon interaction the magnetic impurity mediated electron-electron interaction can play an essential role, which is supported also by new experiments in magnetic field [18]. It has been known since a long time [19] that such interaction is singular in the energy transfer E and recently Kaminski and Glazman [20] called the attention to similar $1/E^2$ singularity in the electron-electron scattering rate phenomenologically suggested by the Saclay group. Using that mechanism the electron transport is determined by the Boltzmann equation and compared with the experimentally determined electron distributions and the impurity concentrations were adjusted [1, 21, 22, 23, 24]. In some cases the estimated magnetic impurity concentrations using that method are much larger than those determined from the dephasing rate, even by two orders of magnitude. In AuPd samples with large spin-orbit interaction the size dependence of dephasing was also observed [14] where for smaller size the electron-electron interaction dominates while for larger samples there are additional scattering mechanism resulting in saturation. That trend is just the opposite what could be expected in case of additional scattering centers at the surface. In the following the possible role of surface anisotropy in these phenomena is discussed.

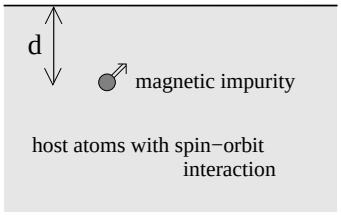

FIG. 1: The magnetic impurity at distance d from the surface.

Surface anisotropy:

For plane-like surfaces (see Fig. 1) the spin-orbit-induced surface anisotropy has the form [2]

$$H = K_d(\mathbf{nS})^2 \tag{1}$$

where $\mathbf{n}$ is the normal direction of the experienced surface element and $\mathbf{S}$ is the spin of the impurity. The anisotropy constant $K_d > 0$ is inversely proportional to the distance d measured from the surface thus it has the form $K_d = \alpha\frac{1}{d}$. For thin films using the assumption that the to surfaces of the film contribute additively, $K_d = \alpha(\frac{1}{d} + \frac{1}{t-d})$, α was obtained from fitting the Kondo resistivity of Au(Fe) and Cu(Fe) films as $\alpha = 247$ÅK [25], of magnetoresistance of Au(Fe) films as $\alpha = 42$ÅK [26], and from multilayer experiments on Au(Fe) films as $\alpha = 60$ÅK [7]. The parameter α depends also on the disorder on the surface and in the bulk.

According to the anisotropy there are different splitting schemes for integer and half-integer spins. For integer spins (e.g. Fe, Cr $S = 2$) the ground state is a singlet, whereas for half-integer spins (e.g. Mn $S = 5/2$) it is a Kramers doublet. Thus for integer spins the anisotropy causes size effects e.g. in Kondo resistivity [3], magnetoresistance [5], thermopower [6], impurity spin magnetization [8], but for half-integer spins not [4, 9, 27]. It is demonstrated that there is a crucial difference between the cases of integer and half-integer spin. That difference can be less pronounced for e.g. $S = 5/2$ as in the spin glass region pairs or clusters can be formed which could have also integer spins showing size dependence [10].

Dephasing:

As the experiments are carried out at low temperature e.g. $T \sim 40$mK, thus in thermal equilibrium most of the higher levels cannot contribute to any dynamics for impurities with large enough anisotropy, $K_d > kT$. In case of integer spins the impurity is frozen in a singlet ground state which cannot lead to dephasing, in contrary to the half-integer spin case where the lowest states form a doublet and we do see dephasing. Samples with Fe implantation or contamination must show a strong size dependence in contrary to Mn.

Energy relaxation:

The nonequilibrium distribution function of a metallic wire with length L and bias U in the diffusive limit is determined by the Boltzmann equation

$$\begin{aligned}\frac{\partial f(\varepsilon,x)}{\partial t} &- \frac{1}{\tau_D}\frac{\partial^2 f(\varepsilon,x)}{\partial^2 x} + I_{coll.}(\{f\}) = 0\\ I_{coll.}(\{f\}) &= \int dE\big\{f(\varepsilon)[1-f(\varepsilon-E)]W(\varepsilon,E)\\ &- [1-f(\varepsilon)]f(\varepsilon-E)W(\varepsilon-E,-E)\big\}\end{aligned} \tag{2}$$

where $W(\varepsilon,E)$ is the scattering rate, $\tau_D = \frac{L^2}{D}$ is the diffusion constant, f is assumed not depending on the spin, and x denotes the position in the wire in the units of L. Starting with the solution without inelastic scattering

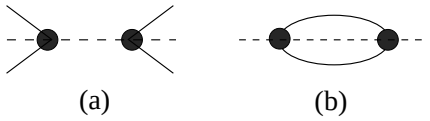

FIG. 2: The diagrams used for calculating (a) the kernel and (b) the Korringa lifetime of the impurity spin. The solid lines denote the conduction electrons, the dotted lines the impurity spin, and the blob is the Kondo coupling.

mechanism

$$f^{(0)}(\varepsilon,x) = (1-x)n_F(\varepsilon - \frac{eU}{2}) + xn_F(\varepsilon + \frac{eU}{2}) \tag{3}$$

and taking into account inelastic scattering in W, the Boltzmann equation can be solved self-consistently at least numerically.

Here we examine the effect of the surface anisotropy on the energy relaxation. Similar to the case of finite magnetic field [1] the first order processes contribute also to the scattering rate and the spin occupation numbers p_Ms depend also on the voltage U. Calculating them from the first order processes we solved the Boltzmann equation self-consistently using the following collision integral

$$\begin{aligned}I^{(2)}_{coll.}(\{f\}) =& \int dE \int d\varepsilon' K^S_{MM'}(E,\varepsilon,\varepsilon',K_d)\big\{p_M f(\varepsilon)f(\varepsilon')\\ &\times[1-f(\varepsilon-E)][1-f(\varepsilon'+E+K_dM^2-K_dM'^2)]\\ &-p_{M'}[1-f(\varepsilon)][1-f(\varepsilon')]f(\varepsilon-E)\\ &\times f(\varepsilon'+E+K_dM^2-K_dM'^2)\big\}\end{aligned} \tag{4}$$

where the kernel $K^S_{MM'}$ describes electron-electron interaction mediated by Kondo impurities with surface anisotropy.

For simplicity we considered the $S = 1$ case when the x-dependent p_Ms are determined from the first order processes by

$$\frac{p_0}{p_1} = \frac{\int d\varepsilon f(\varepsilon,x)(1-f(\varepsilon+K_d,x))}{\int d\varepsilon f(\varepsilon,x)(1-f(\varepsilon-K_d,x))} \tag{5}$$

and $2p_1 + p_0 = 1$.

The kernel in Eq. (4) can be calculated from the diagram Fig. 2 (a) in the Kondo model with anisotropy [25]

$$\begin{aligned}H &= \sum_{k,\sigma}\varepsilon_k\, a^\dagger_{k\sigma}a_{k\sigma} + K_d(\mathbf{nS})^2\\ &+ \sum_{\substack{k,k',\sigma,\sigma'\\ M,M'}} J_{MM'}\mathbf{S}_{MM'}\,(a^\dagger_{k\sigma}\boldsymbol{\sigma}_{\sigma\sigma'}a_{k'\sigma'}),\end{aligned} \tag{6}$$

where $a^\dagger_{k\sigma}$ ($a_{k\sigma}$) creates (annihilates) a conduction electron with momentum k, spin σ and energy ε_k measured from the Fermi level, $\boldsymbol{\sigma}$ stands for the Pauli matrices and $J_{MM'}$'s are the Kondo couplings. The dependence of the

interaction kernel $K^S_{MM'}$ on the energy transfer E for $\tau_K = \infty$ is $(E^2)^{-1}$, $(E \pm K_d)^{-2}$, $(E+K_d)^{-1} \cdot (E-K_d)^{-1}$ in different terms, respectively.

For sake of simplicity we used an appropriate constant value $\tilde{J}$ instead of the renormalized Kondo couplings depending on M, M'. The influence of such an approximation was examined in a preceding self-consistent calculation without surface anisotropy [28]. There the renormalized coupling was calculated as the solution of the leading logarithmic scaling equation assuming similar resummation as in equilibrium and smeared by the spin spectral function with finite Korringa lifetime $\rho_s(\varepsilon) = \frac{1}{\pi} \frac{\frac{\hbar}{2\tau_K}}{\varepsilon^2 + \frac{\hbar^2}{4\tau_K^2}}$. The validity of the logarithmic approximation was always checked by plotting the actual Kondo coupling. From the numerical calculations we can conclude [28] that to get the same results it is a good approximation to replace the renormalized coupling in the kernel by an appropriately chosen constant value. Furthermore, the smearing of the renormalized coupling has very small effect [28] on the results for the parameters consistent with the experimental situation and our results were in complete agreement with Ref.[1].

As the weak dependence on the Korringa lifetime [29] τ_K of the impurity spin we used the value for $K_d = 0$ calculating it from the diagram Fig. 2 (b) as

$$\frac{\hbar}{2\tau_K(x)} = 2\pi(\rho_0\tilde{J})^2 S(S+1) \int d\varepsilon (1 - f(\varepsilon, x)) f(\varepsilon, x). \tag{7}$$

where ρ_0 is the conduction electron density of states for one spin direction.

At each step of the iteration solving the Boltzmann equation self-consistently, both the spin occupation numbers and the Korringa lifetime were calculated from the actual f.

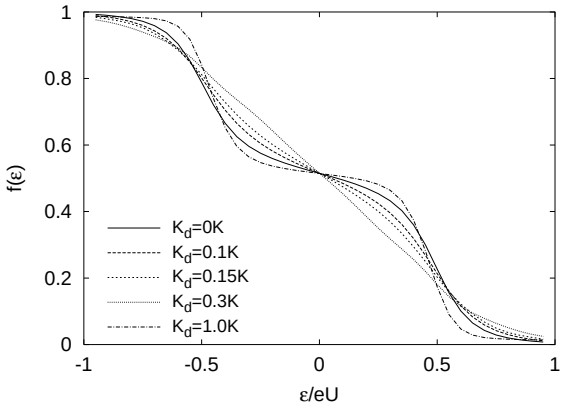

FIG. 3: The calculated distribution function at $x = 0.485$ for different strength of the anisotropy constant K_d. The other parameters are $U = 0.1$mV, $c = 8$ppm, $\rho_0\tilde{J} = 0.11$, and $\tau_D = 2.8$ns.

The dependence of the distribution function on the strength of the anisotropy constant K_d is illustrated in Fig. 3. Increasing K_d first the energy transfer is getting larger but for larger K_d the ground state is frozen in, similar to the magnetic field dependence discussed in Ref. [1]. We can conclude that the contribution of magnetic impurities is enhanced or unchanged in case of finite anisotropy $K_d < eU$. For $K_d \sim 0.1-0.2$K which is a good estimation for the strength of the anisotropy for the wires with width of ~ 45nm and thickness of $\sim 85-110$nm used in the experiments, the energy relaxation is only slightly affected by the anisotropy.

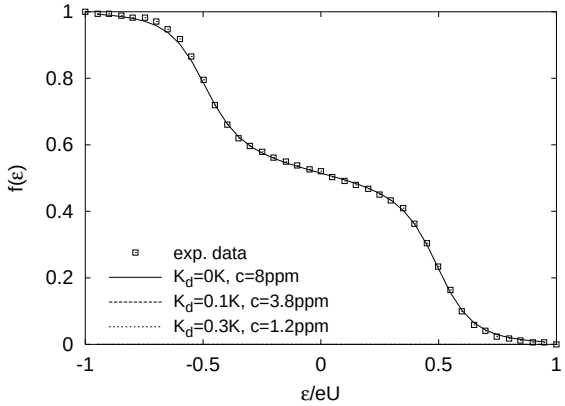

FIG. 4: Fit on the experimental data of Cu wires at $x = 0.485$ [18] by the calculated distribution function for different K_d and c pairs. The other parameters are $U = 0.1$mV, $\rho_0\tilde{J} = 0.11$ and $\tau_D = 2.8$ns.

The goal of the present Letter is not to make optimal fitting of the experimental curves and determine the value of K_d which must have a broad distribution itself. We demonstrate, however, that the experimental curves can be fitted by using combinations of different values of the concentration c and of K_d, and the larger the c is the smaller the necessary K_d. As a demonstration we compare our results to the experimental data on Cu wires at $x = 0.485$ [18] in Fig. 4 as for Cu wires the impurities may be CuO on the surface having $S = 1$ spin [30]. The other fixed parameters are $U = 0.1$mV, $\rho_0\tilde{J} = 0.11$ and $\tau_D = 2.8$ns, and similarly good fits are obtained for $U = 0.3$mV as well. The parameters are somewhat different, which is not surprising as the distribution for K_d is not taken into account. It is important to note, that in some cases the origin of the magnetic impurities is not known, therefore the Kondo temperature corresponding to $\rho_0\tilde{J}$ in our simple approximation is also a fit parameter.

The half-integer case must be very similar to the case without surface anisotropy because of the degeneracy, and only the spin dependent prefactors are different.

The two-level system (TLS) may result in somewhat similar behavior [31]. If $T << \Delta$, where Δ is the splitting, the dephasing is blocked. In the nonequilibrium

case with applied voltage U ($\Delta < eU$) the spin dynamics reenters and could lead to dephasing [32] similarly to the anisotropy case. Similarly, the energy relaxation becomes also possible but to get $\frac{1}{E^2}$ singularity at least two non-commuting couplings are needed [33], thus interaction describing electron screening and electron induced transition between the levels are required [34]. In this case the splitting must be small $\Delta < eU$, but the coupling can be weak enough to be outside the Kondo region. That may result in weak, magnetic field independent contribution, what is suggested by the experiments [35].

In summary, the surface anisotropy for integer spins is suggested to reduce drastically the dephasing rate, while the energy relaxation is much less influenced. In the first case for low temperature and thermal equilibrium the spin dynamics and therefore the dephasing are frozen out while in the out-of equilibrium metallic wire experiments that can reenter. That suggests a pronounced size dependence and very different concentration for the dynamically active impurities in the dephasing and the out-of-equilibrium wire experiments. In the case of half-integer spin having a Kramers doublet as the lowest state instead of a singlet these cannot be expected. Further careful experiments for the size dependence and implanted impurities are required.

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